Sorption Coefficients for Iodine, Silver, and Cesium on Dust Particles

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ABSTRACT

This paper describes the work performed to find relevant experimental data and find the sorption coefficients that represent well the available data for cesium, iodine, and silver on dust particles. The purpose of this work is to generate a set of coefficients that may be recommended for the computer code users. The work was performed using the computer code SPECTRA.

Calculations were performed for the following data:

- I-131 on AVR dust
- Ag-110m on AVR dust
- Cs-13 and Cs-137 on AVR dust

Available data was matched using the SPECTRA Sorption Model.

$$S = A(T) \cdot C_V - B(T) \cdot C_d$$

The results are summarized as follows:

- The available data can be correlated. The data scatter is about 4 orders of magnitude. Therefore the coefficients of the Langmuir isotherms vary by 4 orders of magnitude.
- Sorption rates are higher at low temperatures and lower at high temperatures. This tendency has been observed in the data compiled at Oak Ridge. It is therefore surmised that the highest value of the sorption coefficients are appropriate for the low temperatures and the lowest value of the sorption coefficients are appropriate for the high temperatures. The recommended sorption coefficients are presented in this paper.
- The present set of coefficients is very rough and should be a subject for future verification against experimental data

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1 INTRODUCTION

Sorption of fission product vapors on metallic surfaces and dust particles is an important safety aspect of HTR reactors. Safety analyses of these reactors are performed using computer codes, such as SPECTRA [1], RADAX [2], MELCOR [3]. These codes have sorption models allowing to compute the sorption rates of different fission products on surfaces. The code users must supply the model coefficients applicable for the particular surface and isotope. The sorption process has been theoretically investigated and described in literature [4], [5], [6], [7]. The principal difficulty in applying the sorption models in practice is lack of knowledge of coefficients applicable for each surface and vapor. The coefficients must be based on measured data, which are still quite scarce.

The analysts must often guess values of certain model parameters because the available data is simply insufficient.

Moreover, the available data is sometimes correlated using different coefficients. For example, in case cesium sorption on Incoloy 800, references [6] and [7] show different sets of coefficients based on the Laminar Loop and Vampyr V-II experiments. As shown in reference [1], these different coefficient sets give very different results; the sets that give good results for the Laminar Loop tests fail to represent well the Vampyr test data and vice versa. Data for sorption on dust particles are even more difficult to find than for metallic surfaces.

This paper describes the work performed to find relevant experimental data and find the sorption coefficients that represent well the available data for iodine on dust particles. The purpose of this work is to generate a set of coefficients that may be recommended for the computer code users. The work was performed using the computer code SPECTRA [1].

It is very difficult to find data on sorption on dust particles that would be sufficiently detailed to deduce the sorption coefficients. Some measured data exist from the AVR reactor. This paper presents an effort to correlate the dust activities and the coolant activities, based on the available AVR data. The AVR data has been obtained from reference [7]. The calculations were performed using the SPECTRA code [1]. The SPECTRA sorption correlation was used in such a way as to match the data expressed by Langmuir correlation, which provides equilibrium values of the surface load (mass per unit surface area) of a given isotope for different vapor pressures of this isotope.

The AVR data is shortly discussed in section 2. Section 3 presents results of performed calculations. Finally, section 4 presents conclusions and recommendations.

2 AVR Data

The AVR data was obtained from reference [7]. The data include:

- Coolant activity of isotope *i*, expressed in Bq/Nm^3
- Dust activity of isotope *i*, expressed in Bq/kg

In order to correlate the data by a Langmuir isotherm, the above values need to be converted to:

- Vapor pressure of isotope *i*, expressed in Pa
- Load of isotope *i* on surface, expressed in kg/m^2

Derivation of the conversion formulae is described in reference [12]. The final formulae are shown below.

- Conversion of dust activity, $A_{D,i}$, into surface concentration, $m_{D,i}$:

$$m_{D,i} = \left[\left(\frac{M_i}{N_A \lambda_i} \right) / \left(\frac{6}{\rho_D d_D} \right) \right] \cdot A_{D,i}$$

$m_{D,i}$ surface load of isotope *i* per unit surface area, [kg/m^2]
$A_{D,i}$ activity per unit mass of dust, [Bq/kg]

λ_i	decay constant of the isotope *i*, [1/s]
N_A	Avogadro number (6.02×10^{26} atoms/kmol)
M_i	molar weight of the isotope *i*, [kg/kmol]
d_D,	diameter of dust particle, [m]
ρ_D	density of dust particle, [kg/m^3]:

- Conversion of coolant activity, A_C, into vapor pressure, :

$$p_i = \left[\left(\frac{pM_{He}}{N_A \lambda_i} \right) / \left(\frac{p_N}{R_{He} T_N} \right) \right] \cdot A_{C,i}$$

p_i	partial pressure of vapor of isotope *i*, [Pa]
$A_{C,\,i}$	coolant activity per normal cubic meter, [Bq/Nm^3]
p	total pressure, [Pa]
M_{He}	molar weight of coolant (helium), [kg/kmol]
p_N	normal pressure ($=1.0\times10^5$)
T_N	normal pressure (=273 K)
R_{He}	helium gas constant (equal to 8315/4 = 2080 J/kg K)

3 Results

Several computer codes exist that ara capable of analyzing sorption of fission products on surfaces. These are PATRAS, SPATRA [14], [15], RADAX [2], developed at Jülich, MELCOR [3], developed by Sandia, SPECTRA [1], developed at NRG. The codes have different models for calculating sorption phenomena. In SPECTRA two sorption models are available:

- Sorption Model 1 (SPECTRA model). A simpler model, similar to the one adopted in the MELCOR code.
- Sorption Model 2 (PATRAS/SPATRA model). A more detailed model adopted for the codes PATRAS, SPATRA.

For the present analysis the Sorption Model 1 [1] has been used to represent the available data. The model gives sorption flux as a function of the concentration in the gas phase and on the surface:

$$S = A_S(T) \cdot C_V - B_S(T) \cdot C_d$$

S	sorption mass transfer rate, [kg/(m^2-s)]
$A_S(T)$	adsorption coefficient, [m/s], dependent on wall temperature, T
C_V	concentration of the vapor in the gas space [kg/m^3], with a limit of C_{sat}, $C_V \leq C_{sat}$
$B_S(T)$	desorption coefficient, [1/s]
C_d	concentration of the vapor on the surface, [kg/m^2]

The coefficients $A_S(T)$ and $B_S(T)$ were selected by trial and error to match the available data, as shown in the following sub-sections. The values of $C_{sat}(T)$ were selected to give the saturation activity of about 10^{-6} Bq/m^2 based on the Langmuir isotherm of I-131 on the dust - see [13].

Results are presented in the following three sub-sections:

- Iodine activities - shown in section 3.1.
- Silver activities - shown in section 3.2.
- Cesium activities - shown in section 3.3.

3.1 Iodine Activities

The equilibrium data may be represented by using different combinations of the adsorption and the desorption coefficients. The larger desorption coefficient, $B_S(T)$, is selected, the larger adsorption coefficient, $A_S(T)$, must be used to obtain equilibrium at the desired level. In general the equilibrium is a result of three factors:

- Adsorption, governed by the adsorption coefficient $A_S(T)$
- Desorption, governed by the desorption coefficient $B_S(T)$
- Radioactive decay, governed by the decay constant, λ

In case of iodine the data in reference [6] includes I-131. This isotope has relatively large decay constant ($\lambda = 10^{-6}$ s^{-1}, half-life of $T_{1/2} = 6.9\times10^{5}$ s) and therefore the radioactive decay has a relatively large impact on the equilibrium level.

As a first step a zero desorption coefficient is assumed, $B_S = 0.0$; therefore an equilibrium is a result of interplay between the sorption and the decay. Three adsorption coefficients were selected by trial and error to envelope the measurement data: $A_S = 2.0\times10^{-5}$, 2.0×10^{-3}, 2.0×10^{-1}. Three corresponding values of $C_{sat} = 5.0\times10^{-8}$, 5.0×10^{-10}, 5.0×10^{-12} were selected to give the saturation activity of about 10^{-6} Bq/m^2. Summarizing, the following sorption parameters were assumed:

$$\begin{aligned} A_S(T_w) &= 2.0\times10^{-5} \div 2.0\times10^{-1} \\ B_S(T_w) &= 0.0 \\ C_{sat}(T_w) &= 5.0\times10^{-8} \div 5.0\times10^{-12} \end{aligned}$$

Calculations were performed using a source of isotope and increasing the source strength in steps. After each increase a sufficient time was allowed to reach equilibrium values by both the vapor concentration and the surface concentration on dust particles.

The results are shown Figure 3-1 and Figure 3-2. As seen in Figure 3-1 the steps were sufficiently long to obtain stable conditions at the end of each step. The values of vapor pressure and surface concentration at the end of each step are plotted against each other in Figure 3-2. It is seen that with the selected coefficients the available data is enveloped by the Langmuir isotherm. The fact is that the data spread is about 4 orders of magnitude and therefore such is the spread of the adsorption coefficient. Therefore from the present data one would have to conclude that the data best estimate value of A_S is 2.0×10^{-3}, with quite a large uncertainty, of two orders in magnitude in each direction.

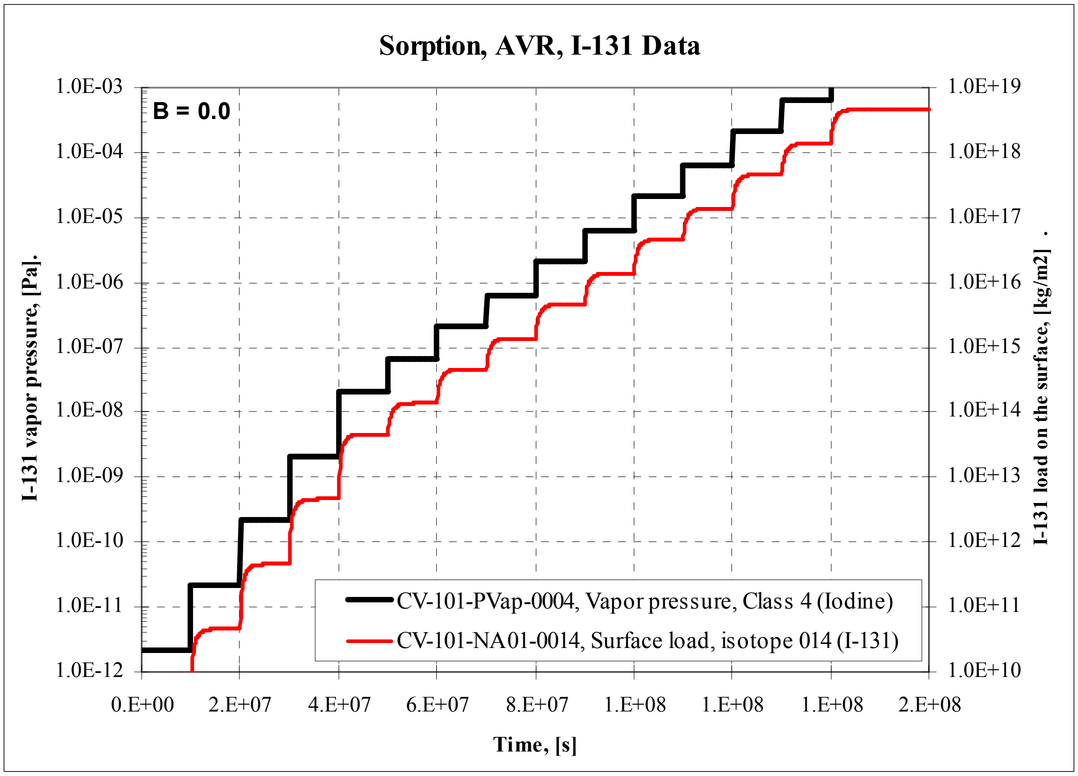

Figure 3-1 I-131 - AVR data and Langmuir isotherms, $B_S = 0.0$

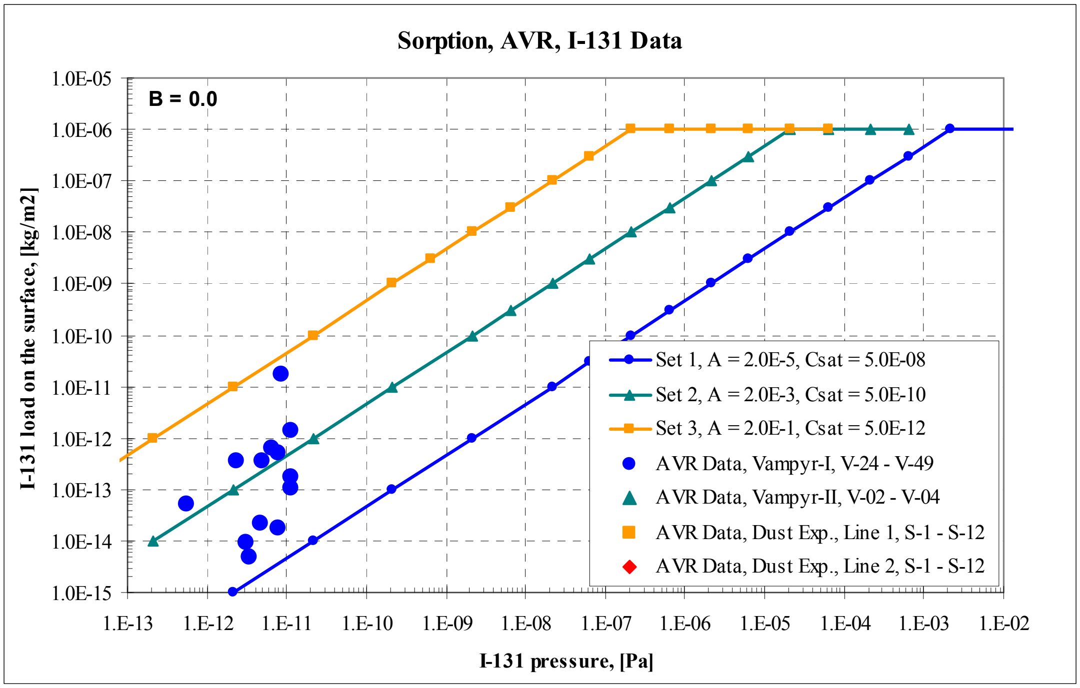

Figure 3-2 I-131 - AVR data and Langmuir isotherms, $B_S = 0.0$

However, based on other data it is expected that the sorption coefficients are higher at low temperatures and lower at high temperatures. This tendency has been observed in the data compiled at Oak Ridge described in reference [5], see also reference [1] Volume 4.

In AVR the dust was continuously circulating, depositing and resuspending. The relative times spend by the dust particles in different parts of the AVR system are not known. It is therefore surmised that the dust particles with the highest activities are those which spent most of their life-time in the relatively cold regions of AVR. This means temperatures of about 200˚C (473 K) - Figure 3-3. On the other hand, the dust particles with the lowest activities are those which spent most of their life-time in the relatively hot regions of AVR. This means temperatures of about 900˚C (1173 K) - Figure 3-3.

If this assumption is correct, it means that the highest value of the sorption coefficients are appropriate for the low temperatures while the lowest value of the sorption coefficients are appropriate for the high temperatures. The data spread (uncertainty) would consequently be much lower than the 4 orders of magnitude.

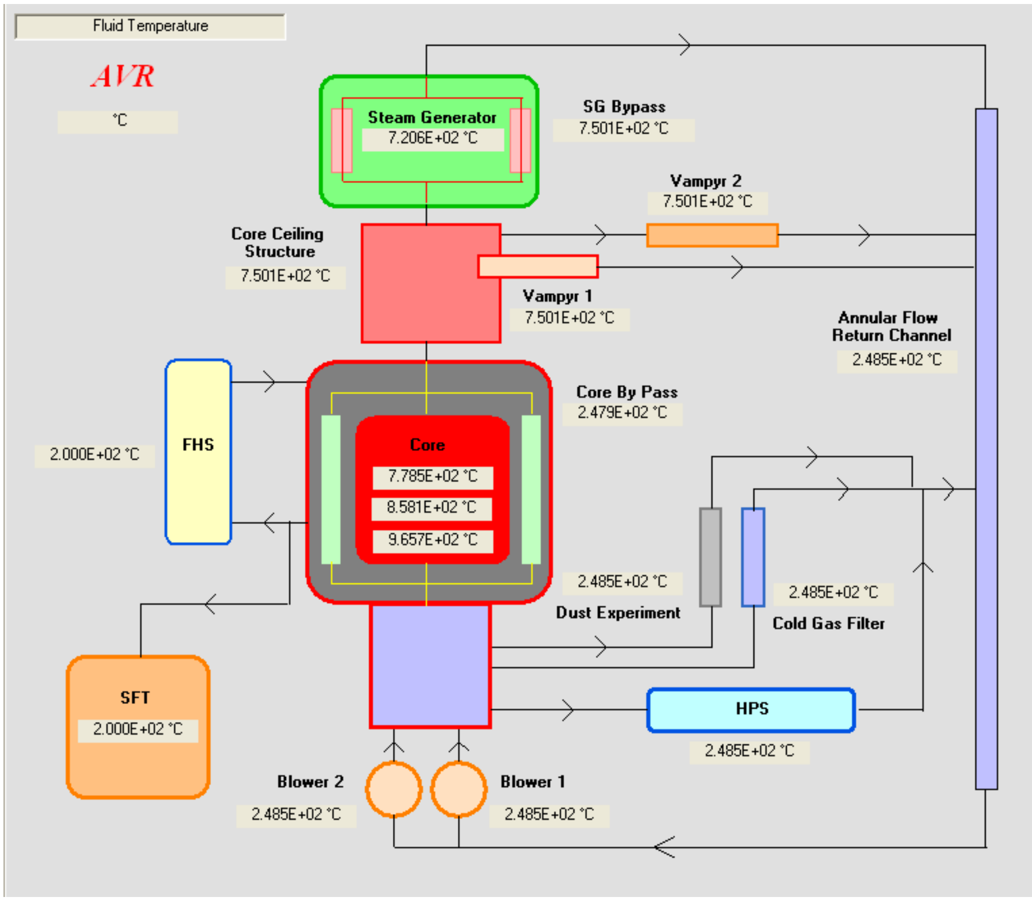

Figure 3-3 **Temperatures in AVR - reference [6]**

Before more detailed measurement data is available this is probably the best working assumption that one can make.

As a next step a desorption coefficient of $B_S = 1.0\times10^{-6}$ was assumed. Again three adsorption coefficients were selected by trial and error to envelope the measurement data: $A_S = 4.5\times10^{-5}$, 4.5×10^{-3}, 4.5×10^{-1}. Three corresponding values of $C_{sat} = 4.0\times10^{-8}$, 4.0×10^{-10}, 4.0×10^{-12} were selected to give the saturation activity of about 10^{-6} Bq/m^2. Summarizing, the following sorption parameters were assumed:

$$\begin{aligned} A_S(T_w) &= 4.5\times10^{-5} \div 4.5\times10^{-1} \\ B_S(T_w) &= 1.0\times10^{-6} \\ C_{sat}(T_w) &= 4.0\times10^{-8} \div 4.0\times10^{-12} \end{aligned}$$

The results are shown in Figure 3-4. The results are very similar to those obtained with $B_S = 0.0$. Thus both combinations (and in fact many other combinations) may be used to represent the available data.

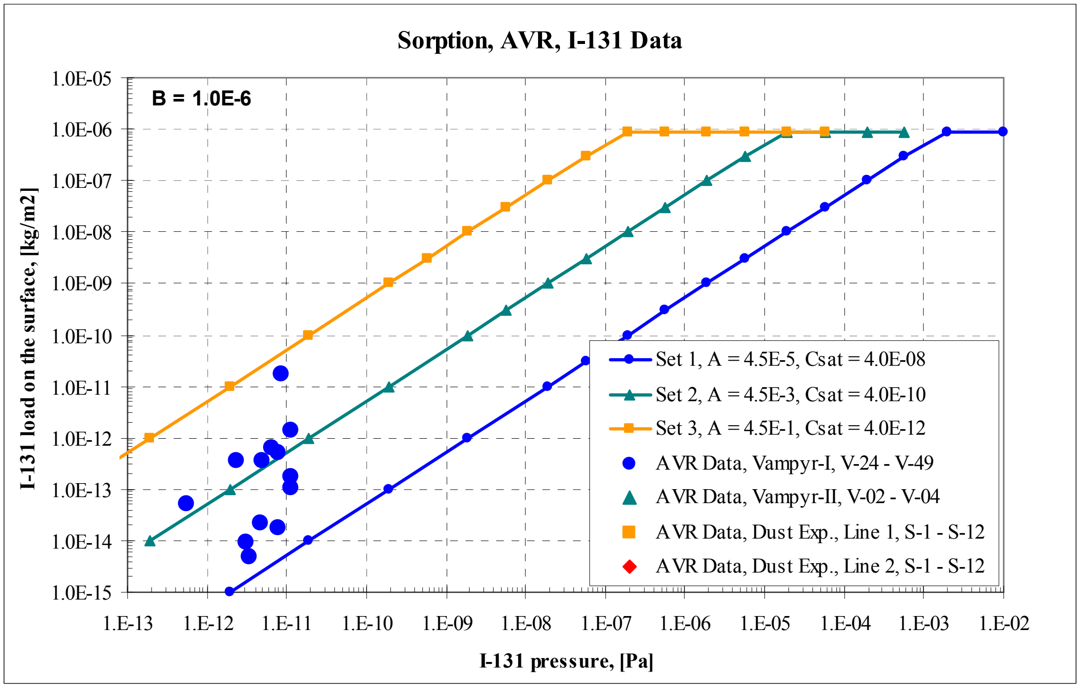

Figure 3-4 I-131 - AVR data and Langmuir isotherms

3.2 Silver Activities

Silver data includes Ag-110m. The vapor class number in SPECTRA is 12. This isotope has relatively small decay constant ($\lambda = 3.21\times10^{-8}$ s^{-1}, half-life of $T_{1/2} = 2.16\times10^{7}$ s) and therefore the radioactive decay has a relatively small impact on the equilibrium level. As a consequence an equilibrium cannot be reached without the desorption coefficient (at least not within a "reasonable" time frame). Therefore the case with $B_S = 0.0$ was not considered. Only $B_S = 10^{-6}$ was used.

Three adsorption coefficients were selected by trial and error to envelope the measurement data: $A_S = 1.0\times10^{-4}$, 1.0×10^{-2}, 1.0. Three corresponding values of $C_{sat} = 1.0\times10^{-8}$, 1.0×10^{-10}, 1.0×10^{-12} were selected to give the saturation activity of about 10^{-6} Bq/m^2. Summarizing, the following sorption parameters were assumed:

$$
\begin{array}{lcll}
A_S(T_w) & = & 1.0\times10^{-4} & \div\ 1.0 \\
B_S(T_w) & = & 1.0\times10^{-6} & \\
C_{sat}(T_w) & = & 1.0\times10^{-8} & \div\ 1.0\times10^{-12}
\end{array}
$$

The results are shown in Figure 3-5.

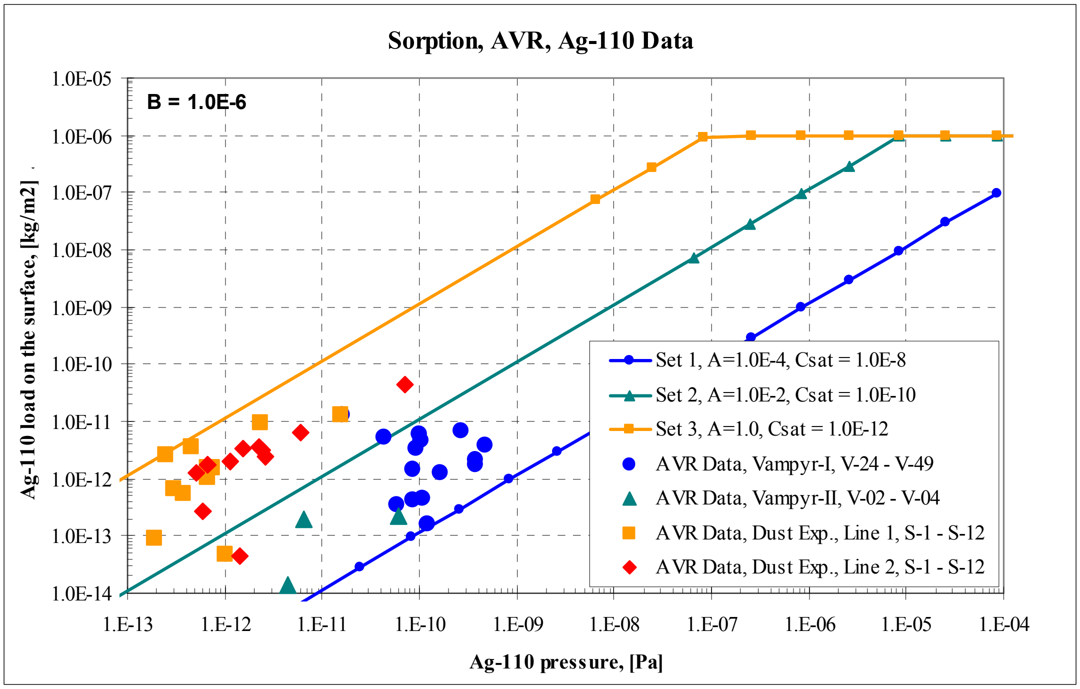

Figure 3-5 **Ag-110m - AVR data and Langmuir isotherms**

3.3 Cesium Activities

Cesium data includes Cs-134 and Cs-137. The vapor class number in SPECTRA is 2. These isotopes have relatively small decay constant ($\lambda = 1.06\times10^{-8}$, 7.33×10^{-10} s^{-1}, half-lives of $T_{1/2}$ = 6.55×10^{7}, 9.5×10^{8} s) and therefore the radioactive decay has very small impact on the equilibrium level. As a consequence an equilibrium cannot be reached without the desorption coefficient. Therefore only the case with $B_S = 10^{-6}$ was used.

Three adsorption coefficients were selected by trial and error to envelope the measurement data: $A_S = 1.0\times10^{-4}$, 1.0×10^{-2}, 1.0. Three corresponding values of $C_{sat} = 1.0\times10^{-9}$, 1.0×10^{-11}, 1.0×10^{-13} were selected to give the saturation activity of about 10^{-6} Bq/m^2. Summarizing, the following sorption parameters were assumed:

$$
\begin{array}{lcll}
A_S(T_w) & = & 1.0\times10^{-4} & \div\ 1.0 \\
B_S(T_w) & = & 1.0\times10^{-6} & \\
C_{sat}(T_w) & = & 1.0\times10^{-8} & \div\ 1.0\times10^{-12}
\end{array}
$$

Results are shown in Figure 3-6.

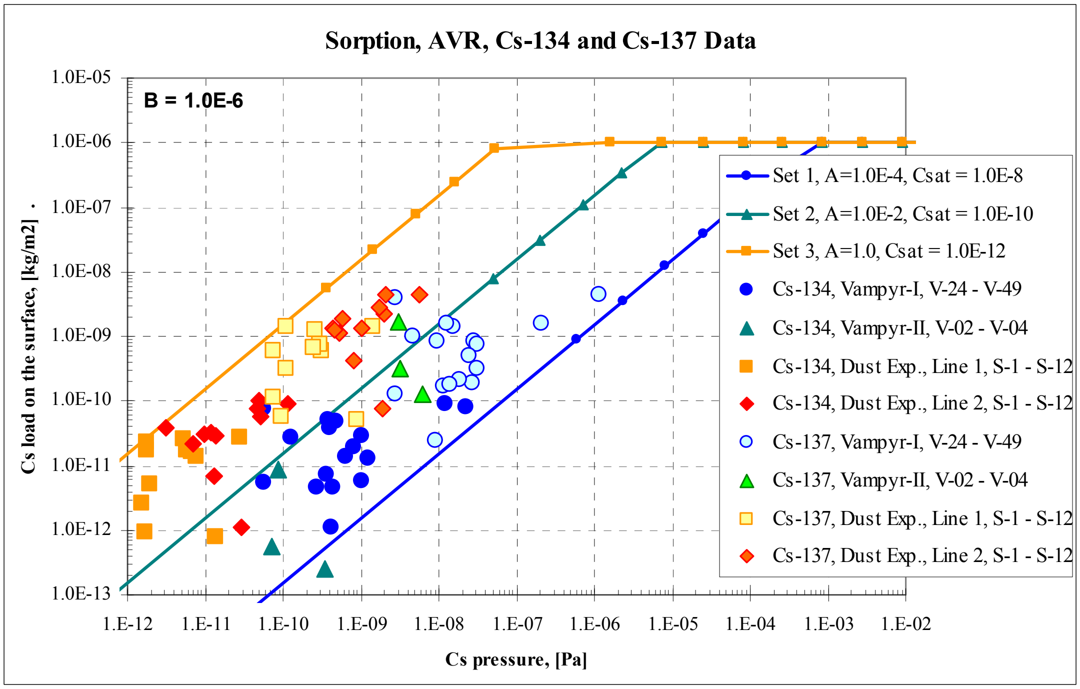

Figure 3-6 **Cs-134 and Cs-137 - AVR data and Langmuir isotherms**

4 CONCLUSIONS AND RECOMMENDATIONS

- The available data can be correlated. The data scatter is about 4 orders of magnitude. Therefore the coefficients of the Langmuir isotherms vary by 4 orders of magnitude.

- Sorption rates are higher at low temperatures and lower at high temperatures. This tendency has been observed in the data compiled at Oak Ridge described in reference [5], see also [13]. It is therefore surmised that the highest value of the sorption coefficients are appropriate for the low temperatures and the lowest value of the sorption coefficients are appropriate for the high temperatures. The recommended sorption coefficients are:

  - Iodine on dust.

$$A_S(T_w) = \begin{cases} 4.5\times10^{-1} & low\ T\ (T \sim 400-500\ K) \\ 4.5\times10^{-3} & middle\ T\ (T \sim 700-800\ K) \\ 4.5\times10^{-5} & high\ T\ (T \sim 1000-1100\ K) \end{cases}$$

$$B_S(T_w) = 1.0\times10^{-6}$$

$$C_{sat}(T_w) = \begin{cases} 4.0\times10^{-12} & low\ T\ (T \sim 400-500\ K) \\ 4.0\times10^{-10} & middle\ T\ (T \sim 700-800\ K) \\ 4.0\times10^{-8} & high\ T\ (T \sim 1000-1100\ K) \end{cases}$$

  - Silver on dust:

$$A_S(T_w) = \begin{cases} 1.0 & low\, T\ (T \sim 400-500\ K) \\ 1.0\times10^{-2} & middle\, T\ (T \sim 700-800\ K) \\ 1.0\times10^{-4} & high\, T\ (T \sim 1000-1100\ K) \end{cases}$$

$$B_S(T_w) = 1.0\times10^{-6}$$

$$C_{sat}(T_w) = \begin{cases} 1.0\times10^{-12} & low\, T\ (T \sim 400-500\ K) \\ 1.0\times10^{-10} & middle\, T\ (T \sim 700-800\ K) \\ 1.0\times10^{-8} & high\, T\ (T \sim 1000-1100\ K) \end{cases}$$

- Cesium on dust.

$$A_S(T_w) = \begin{cases} 1.0 & low\, T\ (T \sim 400-500\ K) \\ 1.0\times10^{-2} & middle\, T\ (T \sim 700-800\ K) \\ 1.0\times10^{-4} & high\, T\ (T \sim 1000-1100\ K) \end{cases}$$

$$B_S(T_w) = 1.0\times10^{-6}$$

$$C_{sat}(T_w) = \begin{cases} 1.0\times10^{-12} & low\, T\ (T \sim 400-500\ K) \\ 1.0\times10^{-10} & middle\, T\ (T \sim 700-800\ K) \\ 1.0\times10^{-8} & high\, T\ (T \sim 1000-1100\ K) \end{cases}$$

- To be on the conservative side (higher sorption) one should apply the coefficients at the higher temperature of the mentioned temperature range (i.e. 500, 800, 1100 K).

- The present set of coefficients is very rough and should be a subject for future verification against experimental data.

- The coefficients presented here are of course only applicable for the particular sorption model, one of several sorption models available in different codes.

Nomenclature

Symbols

$A(T)$, $B(T)$	temperature-dependent sorption coefficients
$A_{C,\,i}$	coolant activity per normal cubic meter, [Bq/Nm^3]
$A_{D,\,i}$	activity per unit mass of dust, [Bq/kg]
C_d	isotope concentration on the surface [kg/m^2]
C_S,	surface load, [mol/m^2]
C_V	isotope concentration in the gas volume [kg/m^3]
d_D,	diameter of dust particle, [m]
$m_{D,\,i}$	surface load of isotope *i* per unit surface area, [kg/m^2]
M_{He}	molar weight of coolant (helium), [kg/kmol]
M_i	molar weight of the isotope *i*, [kg/kmol]
N_A	Avogadro number (6.02×10^{26} atoms/kmol)
p	total pressure, [Pa]
p_i	partial pressure of vapor of isotope *i*, [Pa]
p_N	normal pressure ($=1.0\times10^5$)
R_{He}	helium gas constant (equal to 8315/4 = 2080 J/kg K)
S	sorption flux, [kg/m^{2}s]
T_N	normal pressure (=273 K)

Greek symbols

λ_i	decay constant of the isotope *i*, [1/s]
ρ_D	density of dust particle, [kg/m^3]:

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